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DTU Nutech Report

Sven P. Nielsen, Kasper G. Andersson and Arne Miller
DTU-Nutech-16(EN)rev
December 2017

DTU Nutech
Center for Nuclear Technologies

Author: Sven P. Nielsen, Kasper G. Andersson and Arne Miller
Title: Radioactivity in the Risø District January-June 2017
Center for Nuclear Technologies

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Abstract (max. 2000 char.): The environmental surveillance of the Risø environment was continued in January-June 2017. The mean concentrations in air were: 0.29±0.28 μBq m^{-3} of ^{137}Cs, 2.92±0.92 mBq m^{-3} of ^{7}Be and 0.27±0.23 mBq m^{-3} of ^{210}Pb (±1 S.D.). The depositions by precipitation at Risø in the first half of 2017 were: 0.037±0.006 Bq m^{-2} of ^{137}Cs, 608±61 Bq m^{-2} of ^{7}Be, 12.7±1.1 Bq m^{-2} of ^{210}Pb and <0.9 kBq m^{-2} of ^{3}H. The average background dose rate (TLD) at Risø (Zone I) was measured as 51 nSv h^{-1} compared with 48 ± 3 nSv h^{-1} (±1 S.D.) in the four zones around Risø.

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INTRODUCTION

A specific monitoring programme in the vicinity of the nuclear installations at the Risø site is carried out by DTU Nutech on behalf of and as a contractor to Danish Decommissioning (DD). This report presents the analytical results of the monitoring and sampling carried out in the period January-June 2017. The materials and methods used in connection with the monitoring programme are described in pages 23-24.

Table 1. Radionuclides in ground level air collected at Risø (cf. Figs. 1, 1.1 and 1.2), January - June 2017 (Unit: $\mu Bq\ m^{-3}$)

Date			^{7}Be	^{137}Cs	^{210}Pb
03-Jan-17	–	10-Jan-17	2740(10%)*	0.353(12%)	232(10%)
10-Jan-17	–	17-Jan-17	2251(10%)	0.337(13%)	193(10%)
17-Jan-17	–	24-Jan-17	1696(10%)	0.246(15%)	183(10%)
24-Jan-17	–	31-Jan-17	4055(10%)	0.593(12%)[a]	526(10%)
31-Jan-17	–	06-Feb-17	2915(10%)	0.840(12%)[a]	1229(10%)
06-Feb-17	–	13-Feb-17	3946(10%)	0.853(10%)[a]	513(10%)
13-Feb-17	–	20-Feb-17	3218(10%)	1.175(11%)[a]	513(10%)
20-Feb-17	–	27-Feb-17	1409(10%)	0.097(29%)	67(10%)
27-Feb-17	–	07-Mar-17	2561(10%)	0.175(17%)	213(10%)
07-Mar-17	–	14-Mar-17	1688(10%)	0.202(12%)	242(10%)
14-Mar-17	–	22-Mar-17	2725(10%)	0.198(11%)	86(10%)
22-Mar-17	–	28-Mar-17	2167(10%)	0.267(11%)	112(10%)
28-Mar-17	–	03-Apr-17	2183(10%)	0.199(12%)	226(10%)
03-Apr-17	–	10-Apr-17	1874(10%)	0.150(13%)	89(10%)
10-Apr-17	–	18-Apr-17	2215(10%)	0.128(11%)	151(10%)
18-Apr-17	–	24-Apr-17	3805(10%)	0.199(12%)	188(10%)
24-Apr-17	–	01-May-17	2840(10%)	0.174(11%)	138(10%)
01-May-17	–	08-May-17	4349(10%)	0.413(11%)	296(10%)
08-May-17	–	15-May-17	4307(10%)	0.227(12%)	256(10%)
15-May-17	–	22-May-17	4918(10%)	0.206(13%)	350(10%)
22-May-17	–	29-May-17	3435(10%)	0.118(13%)	295(10%)
29-May-17	–	06-Jun-17	3340(10%)	0.099(16%)	215(10%)
06-Jun-17	–	12-Jun-17	3552(10%)	0.055(13%)	166(10%)
12-Jun-17	–	19-Jun-17	2271(10%)	0.050(15%)	163(10%)
19-Jun-17	–	26-Jun-17	2593(10%)	0.050(15%)	154(10%)
26-Jun-17	–	04-Jul-17	2944(10%)	0.111(12%)	134(10%)
Mean			2923	0.289	267
SD			921	0.279	233

*Figures in brackets are relative standard uncertainties

[a] Increased concentrations possibly caused by forest fires in areas of Eastern Europe contaminated from the Chernobyl accident.

Table 2.1. Radionuclides in precipitation in the 10 m^2 rain collector at Risø (cf. Fig. 8.1), January - June 2017. (Unit: Bq m^{-3})

Month	^{7}Be	^{137}Cs	^{210}Pb
January	1841(10%)*	0.262(44%)	243(11%)
February	1388(10%)	0.077(35%)	106(10%)
March	1140(10%)	0.091(26%)	74(11%)
April	669(10%)	0.069(21%)	49(10%)
May	3155(10%)	0.917(37%)[a]	183(17%)
June	1904(10%)	0.120(28%)	101(11%)

*Figures in brackets are relative standard uncertainties

[a]Note: Enhanced concentration due to low precipitation in this month, see Table 2.2.

Table 2.2. Radionuclides in precipitation in the 10 m^2 rain collector at Risø (cf. Fig. 8.1), January - June 2017. (Unit: Bq m^{-2})

Month	Precipitation (m)	^{7}Be	^{137}Cs	^{210}Pb
January	0.035(10%)*	13.6(14%)	0.0019(46%)	1.8(14%)
February	0.039(10%)	49.9(14%)	0.0028(37%)	3.8(15%)
March	0.041(10%)	44.8(14%)	0.0036(28%)	2.9(14%)
April	0.061(10%)	40.7(14%)	0.0042(21%)	3.0(14%)
May	0.012(10%)	39.0(14%)	0.0113(39%)	0.2(14%)
June	0.113 (10%)	217.0(14%)	0.0136(29%)	1.0(12%)
Sum	0.301(5%)	608.4(10%)	0.0374(15%)	12.7(9%)

*Figures in brackets are relative standard uncertainties

Table 2.3. Tritium in precipitation collected at Risø (cf. Figs. 1, 2.3.1 and 2.3.2). January - June 2017. (Unit: kBq m^{-3})

Month	10 m^2 rain collector*
January	5.3(16%)[a]
February	2.9(27%)
March	3.4(27%)
April	< 2.2
May	< 2.2
June	< 2.2
Double determinations*.	

[a] Figures in brackets are relative standard uncertainties

Table 2.4. Tritium in precipitation collected at Risø (cf. Fig. 1). January - June 2017. (Unit: kBq m^{-2})

Month	Precipitation (m)	10 m^2 rain collector
January	0.035(10%)[a]	0.185(19%)
February	0.039(10%)	0.113(29%)
March	0.041(10%)	0.139(29%)
April	0.061(10%)	< 0.135
May	0.012(10%)	< 0.027
June	0.113 (10%)	< 0.249
Sum	0.301(5%)	< 0.848

[a] Figures in brackets are relative standard uncertainties

Table 3.1. Radionuclides in sediment samples collected at Bolund in Roskilde Fjord.(cf. Fig. 3.1) January - June 2017. (Unit: Bq kg^{-1} dry)

No samples in this period.

Table 4.1. Radionuclides in seawater collected in Roskilde Fjord (cf. Fig. 4.1) January - June 2017. (Unit: Bq m^{-3})

No samples in this period.

Table 4.2. Tritium in seawater collected in Roskilde Fjord (Risø pier) (cf. Fig. 4.2) January - June 2017.

Month	kBq m^{-3}
March	< 2.2 *
June	< 2.2

* Double determinations

.

Table 5.1. Radionuclides in grass (snow) collected at Risø near the Waste Treatment Station, location I P3, Fig. 1, January - June 2017. (**Measured on bulked ash samples)*

Week no. or month	Date	K (g kg^{-1} fresh)	^{137}Cs (Bq kg^{-1} fresh)	^{137}Cs (Bq m^{-2})
1	2 January	2.1(11%)[a]	<0.3	
3	17 January	2.1(11%)	<0.3	
5	31 January	2.4(11%)	<0.4	
7	13 February	2.4(11%)	<0.2	
9	27 February	4.1(10%)	<0.2	
11	14 March	3.6(11%)	<0.6	
13	28 March	3.4(11%)	<0.7	
15	10 April	5.2(11%)	<0.6	
17	24 April	3.0(10%)	<0.3	
19	8 May	4.6(13%)	<1.5	
21	22 May	5.5(10%)	<0.4	
23	6 June	5.7(10%)	<0.3	
25	19 June	5.6(10%)	<0.4	
**January		2.2(10%)	0.159(13%)	0.077(16%)
**February		3.1(10%)	0.662(11%)	0.037(14%)
**March		3.6(10%)	0.217(23%)	0.048(26%)
**April		3.7(10%)	0.156(24%)	0.050(27%)
**May		5.9(10%)	0.059(23%)	0.012(26%)
**June		6.4(10%)	0.053(25%)	0.032(28%)

[a] Figures in brackets are relative standard uncertainties

Table 5.2. Radionuclides in Fucus vesiculosus collected at Bolund in Roskilde Fjord. January - June 2017. (Unit: Bq kg^{-1} dry)

No samples in this period.

Table 7.1. Waste water collected at Risø (cf. Fig. 1), January - June 2017.

Week number	eqv. mg KCl l^{-1}	^{137}Cs (Bq m^{-3})	^{131}I (Bq m^{-3})	^{226}Ra (Bq m^{-3})
1	81(11%)*	<122	<139	<298
2	87(11%)	<133	<126	<287
3	63(13%)	<124	<134	<269
4	83(11%)	<127	<135	<303
5	85(10%)	<120	<122	<287
6	70(15%)	<120	<138	<288
7	81(13%)	<137	<135	<318
8	71(11%)	<144	<152	<359
9	55(12%)	<122	<142	<292
10	80(11%)	<124	<134	<289
11	75(11%)	<124	<125	<293
12	61(11%)	<130	<134	<308
13	57(10%)	<114	<124	<271
14	67(11%)	<119	<131	<289
15	79(10%)	<116	<125	<289
16	61(11%)	<118	<144	<289
17	82(11%)	<127	<131	<293
18	44(16%)	<128	<138	<312
19	45(14%)	<130	<142	<328
20	70(10%)	<110	<123	<301
21	100(11%)	<137	<146	<348
22	98(11%)	<130	<132	<301
23	94(11%)	<135	<139	<337
24	76(11%)	<127	<123	<294
25	86(10%)	<132	<139	<321
26	73(10%)	<120	<128	<290
Mean	74.0	<126	<134	<302
SD	14.7			

* Figures in brackets are relative standard uncertainties

Table 8.1. Background dose rates around the border of Risø (cf. Fig. 8.1) measured with thermoluminescence dosimeters (TLD) in the period November 2016 – April 2017. (Results are normalized to nSv h^{-1})

Location	nSv h^{-1} ¤
1	41(10%)[a]
2	40(10%)
3	51(10%)
4	50(10%)
5	47(10%)
6	53(10%)
Mean	47(5%)

[a] Figures in brackets in Table 8.1 and 8.2 are relative standard uncertainties

Table 8.2. Background dose rates around Risø (cf. Fig. 8.2 and Fig. 1) measured with thermoluminescence dosimeters (TLD) in the period November 2016– April 2017. (Results are normalized to nSv h^{-1}),

Risø zone	Location	nSv h^{-1} ¤
I	1	39(10%)[a]
I	2	47(10%)
I	3	68(10%)
I	4	48(10%)
I	5	55(10%)
Mean		51(5%)
II	P1	54(10%)
II	P2	51(10%)
II	P3	40(10%)
II	P4	Dosimeter lost
Mean		48(10%)
III	P1	45(10%)
III	P2	47(10%)
III	P3	42(10%)
Mean		45(6%)
IV	P1	36(10%)
IV	P2	39(10%)
IV	P3	50(10%)
IV	P4	46(10%)
IV	P5	50(10%)
IV	P6	45(10%)
IV	P7	57(10%)
Mean		46(4%)
V	P1	54(10%)
V	P2	59(10%)
V	P3	57(10%)
V	P4	41(10%)
V	P5	50(10%)
V	P6	42(10%)
V	P7	44(10%)
V	P8	54(10%)
V	P9	46(10%)
V	P10	58(10%)
Mean		51(4%)

Table 8.3. Terrestrial dose rates at the Risø zones (cf. Fig. 8.2 and Fig. 1) January – June 2017. Measured with a NaI(Tl) detector. (Unit: nSv h^{-1})

Risø zone	Location	October
I	P1	46(10%)[a]
I	P2	52(10%)
I	P3	326(10%)
I	P4	47(10%)
I	P5	49(10%)
Mean		104(5%)
II	P1	46(10%)
II	P2	46(10%)
II	P3	43(10%)
II	P4	41(10%)
Mean		44(4%)
III	P1	45(10%)
III	P2	46(10%)
III	P3	42(10%)
Mean		45(6%)
IV	P1	38(10%)
IV	P2	48(10%)
IV	P3	45(10%)
IV	P4	42(10%)
IV	P5	35(10%)
IV	P6	44(10%)
IV	P7	48(10%)
Mean		43(4%)
V	P1	61(10%)
V	P2	54(10%)
V	P3	57(10%)
V	P4	51(10%)
V	P5	51(10%)
V	P6	54(10%)
V	P7	44(10%)
V	P7a	38(10%)
V	P8	49(10%)
V	P9	54(10%)
V	P10	42(10%)
Mean		51(4%)

[a] Figures in brackets are relative standard uncertainties

Fig. 1. Locations for measurements of gamma-background radiation Zone I and II (cf. Tables 8.2 and 8.3)

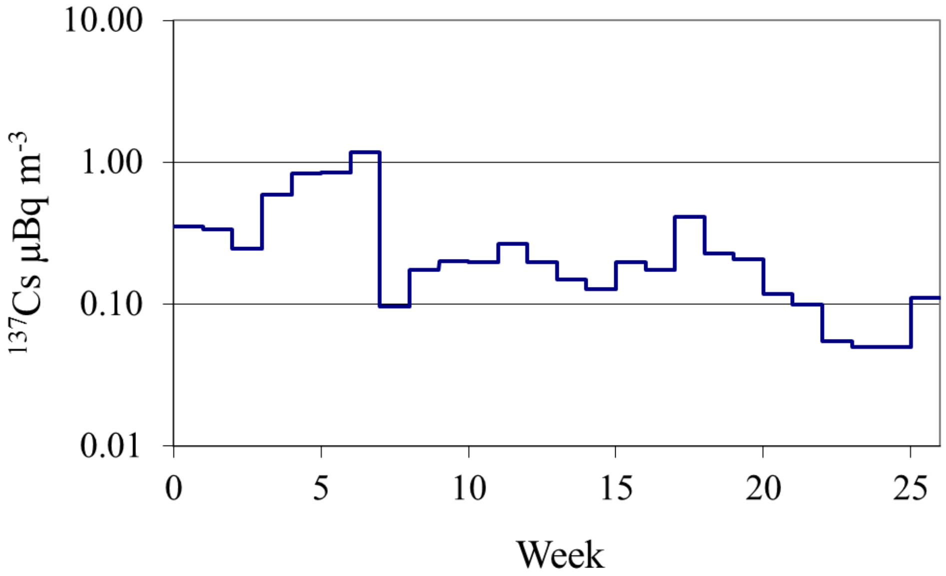

Fig. 1.1. Caesium-137 in ground level air collected at Risø in January-June 2017. (Unit: µBq m^{-3})

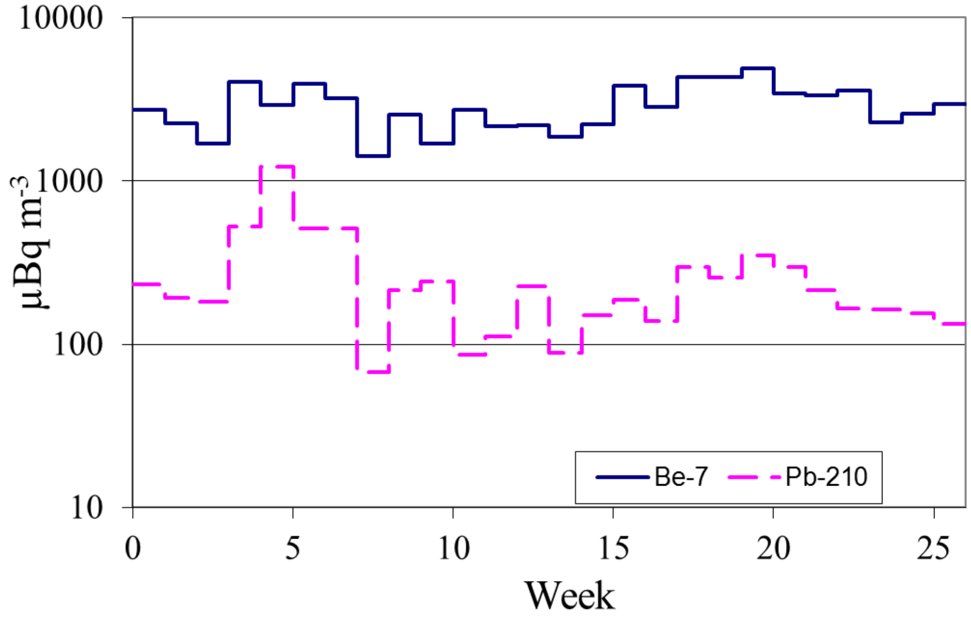

Fig. 1.2. Beryllium-7 and Lead-210 in ground level air collected at Risø in January-June 2017. (Unit: µBq m^{-3})

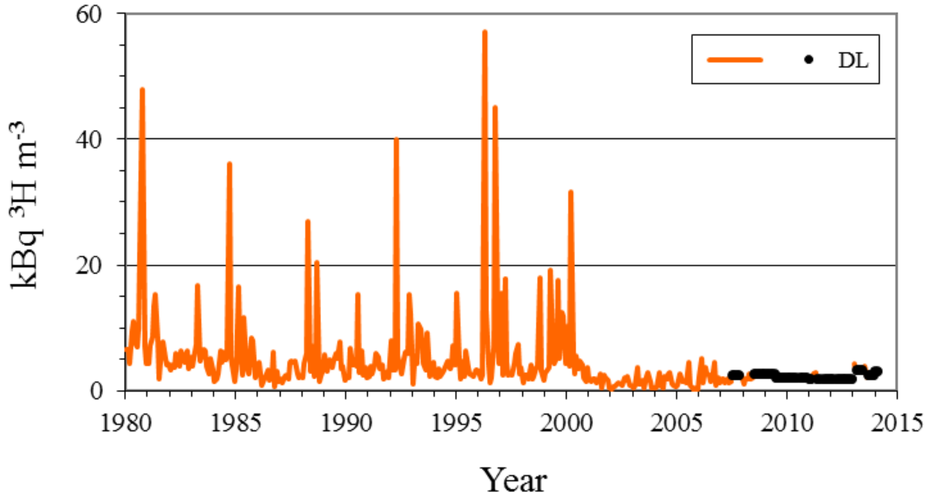

Fig. 2.3.1. Tritium in precipitation collected at Risø (1 m^2 rain collector) 1980 - 2013. (Unit: kBq m^{-3}; DL = detection limit . This rain collector was taken out of operation in 2013.

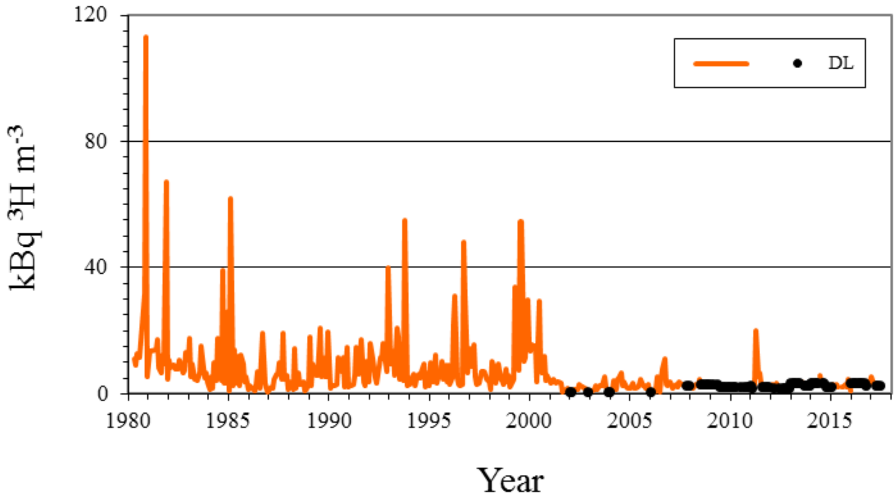

Fig. 2.3.2. Tritium in precipitation collected at Risø (10 m^2 rain collector) 1980 - 2017. (Unit: kBq m^{-3}; DL = detection limit)

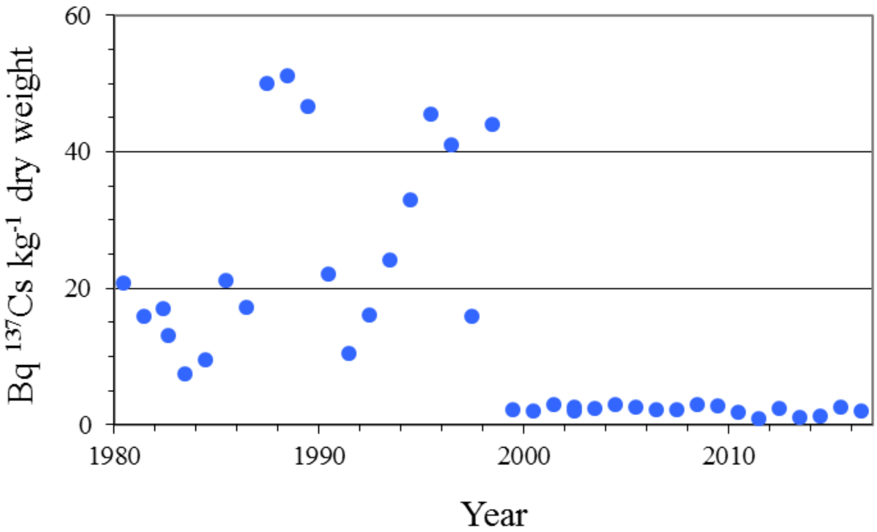

Fig. 3.1. Caesium–137 in sediment samples collected at Bolund in Roskilde Fjord. 1980 – 2017. (Unit: Bq kg^{-1} dry matter)

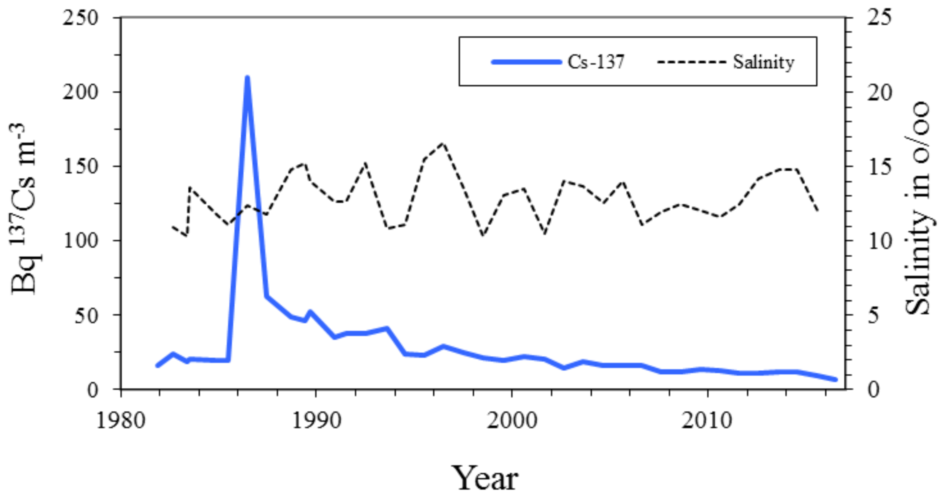

Fig. 4.1. Caesium–137 in seawater collected in Roskilde Fjord 1980 - 2017. (Unit: Bq m^{-3})

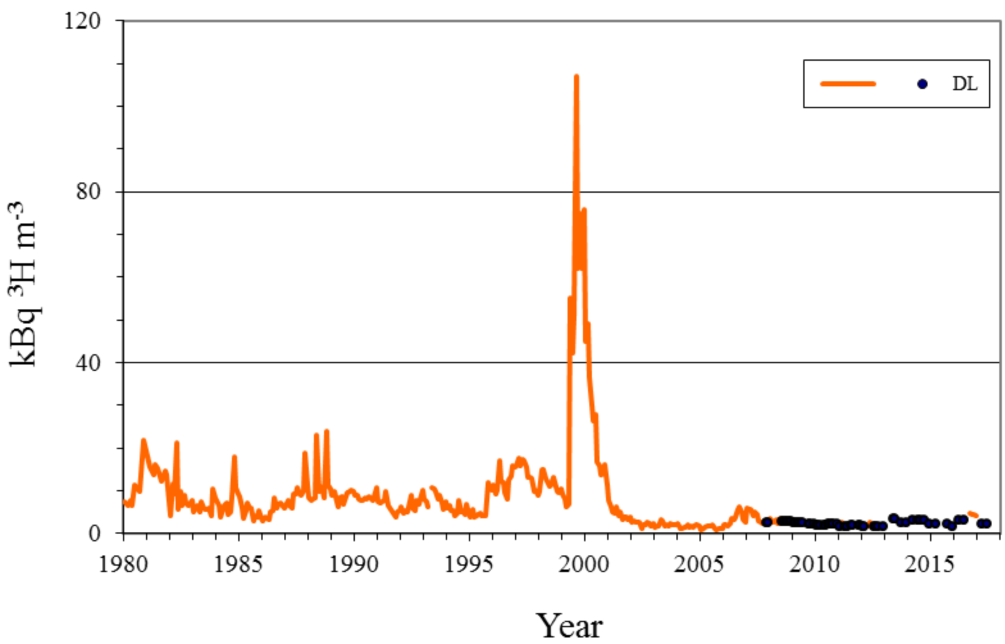

Fig. 4.2. Tritium in seawater collected in Roskilde Fjord 1980 - 2017. (Unit: kBq m^{-3}; DL = detection limit)

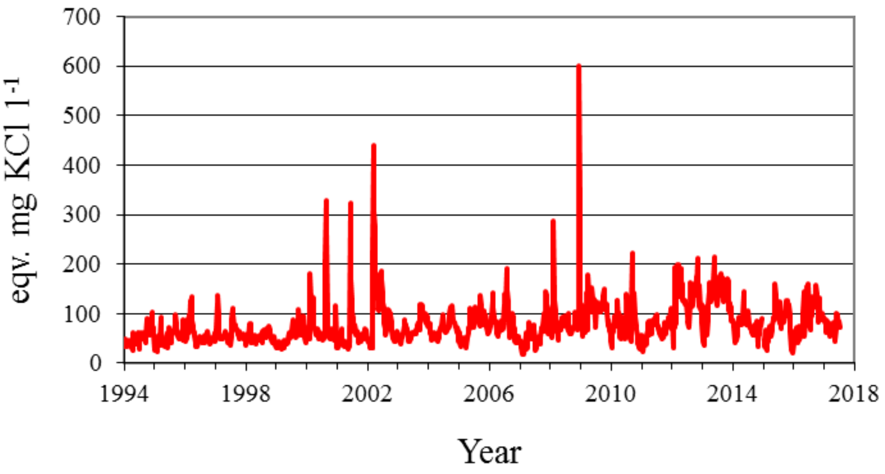

Fig. 7.1. Total-beta radioactivity in waste water collected at Risø 1994 - 2017. (Unit: eqv. mg KCl l^{-1})

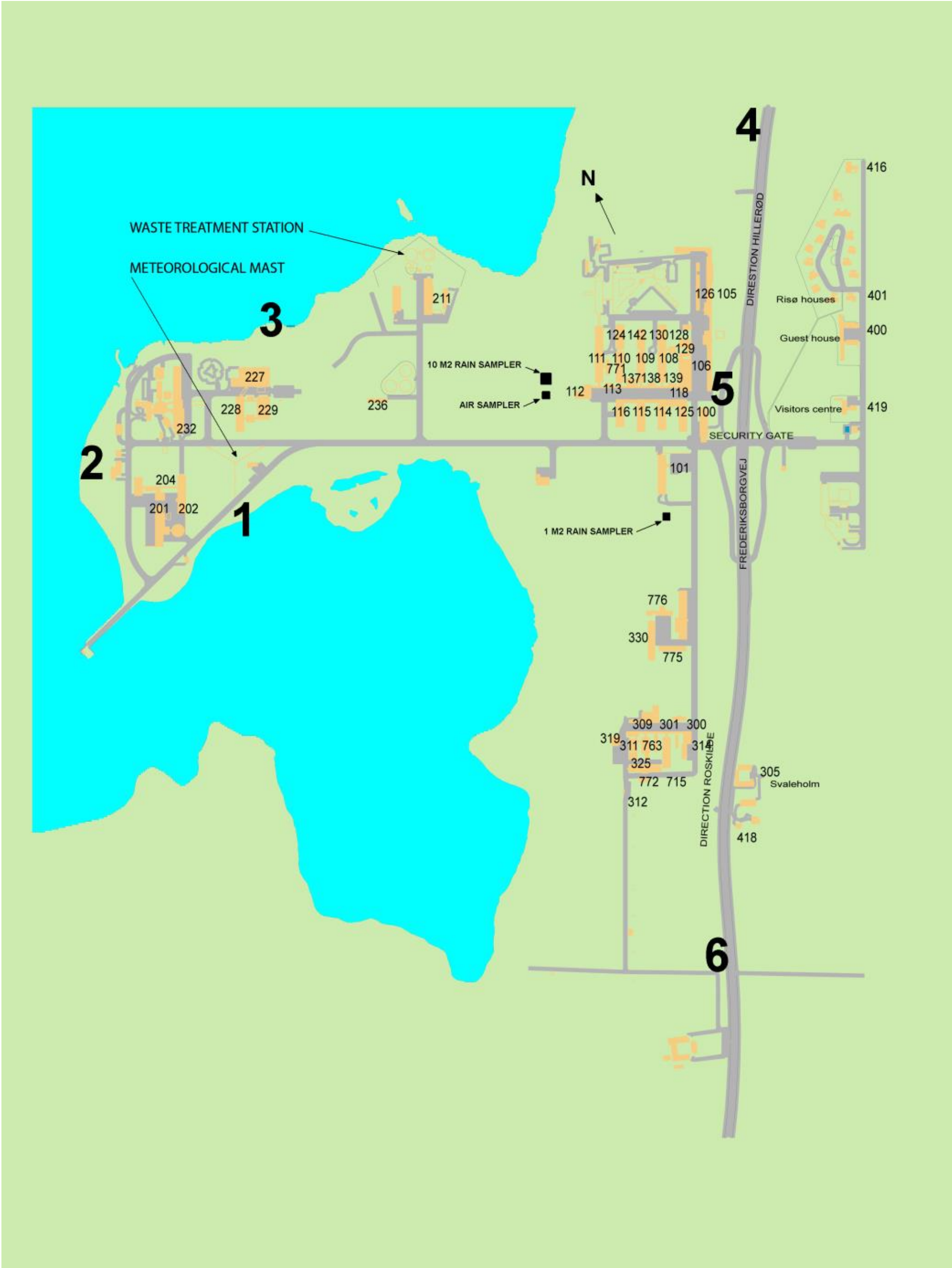

Fig. 8.1. Locations (1-6) for TLD measurements around the border of Risø (cf. Table 8.1).

Fig. 8.2. Locations for measurements of background radiation around Risø in Zones III, IV and V.

MATERIALS AND METHODS

External gamma dose rate monitoring

Monitoring of external gamma dose rate is carried out with the following devices

- Thermolumiscence dosimeters TLD: LiF, measurement frequency annually from May to April. TLD equipment manufacturer: ALNOR/RADOS
- NaI detector: 3x3 inch, SAM 935 Surveillance and Measurement System, Berkeley Nucleonics Cooperation, USA, visual read-out

Calibration of TLD is carried out by irradiation of dosimeters at a calibration irradiator. Traceability of delivered doses is ensured through calibration of the dose rate of the calibration irradiator by the National Institute of Radiation Protection (SIS). Calibration has been verified by measurement with ionisation chamber from NPL, UK. The NaI detector is calibrated periodically vs. a Reuter Stokes high-pressure ionisation chamber.

Air sampler

The sampler at Risø is manufactured by DTU. Air is drawn through a polypropylene filter at a rate of about 2000 m^3/h. The filter is normally changed weekly. The flow rate is monitored by a gas meter connected to a shunt. The gas meter reading is compared to that of a reference gas meter intermittently.

DTU analyse the filters by gamma spectrometry shortly after filter change to check for the presence of short-lived man-made radionuclides. The air filters are subsequently stored for a minimum of one week to allow for decay of short-lived naturally occurring radionuclides before repeated gamma analysis. Filters are analysed for ^{137}Cs, ^{7}Be and ^{210}Pb and other gamma emitters.

Deposition collector

The Risø site operates a large rain collector of 10 m^2. The collector is heated and water is passed through an ion exchange column to a large tank. The 10 m^2 collector provides monthly samples of rain water analysed for tritium and ion exchange resin which is analysed by gamma spectrometry for ^{7}Be, ^{137}Cs and ^{210}Pb and other gamma emitters.

Water and sediment

A waste water sample from the Waste Treatment Station is collected weekly and analysed for total beta radioactivity and the radionuclides ^{131}I, ^{137}Cs and ^{226}Ra. Water samples from Roskilde Fjord are collected each quarter and analysed for tritium, annually for ^{137}Cs. A sediment sample is collected annually from Roskilde Fjord and analysed for ^{137}Cs.

Terrestrial and aquatic biota and flora

Grass samples are collected weekly at the Risø site and analysed by gamma spectrometry. Samples are bulked to monthly samples which are analysed for ^{137}Cs.

Seaweed samples are collected annually from Roskilde Fjord at Risø and analysed for ^{137}Cs.

Sample reception and preparation

Sample identification numbers are entered in log books. Sample preparation methods include drying, freeze drying, ashing, sorting and sieving. Selected samples are archived.

Sample measurements

Radioactivity in samples is measured by total beta counting and gamma spectrometry.

Measurement devices

- Ge detectors for gamma spectrometry. Calibration of detectors is based on mixed-nuclide standards used occasionally. Monthly checks are made of detector efficiency and energy resolution. Background measurements of gamma systems are made a few times per year.
- Low-level Geiger-Müller counters for total beta counting, manufactured by DTU. Calibration based on standards of KCl. Counting efficiency and background are checked monthly.
- Liquid scintillation spectrometer for analysis of tritium in water. Samples are analysed with a calibration standard.

Analytical results, data handling and reporting tools

Analytical results are printed on paper, recorded in log books and stored in a data base on intranet. Results below detection limits recorded as such. Spreadsheets are used for calculating results from raw data.

Quality assurance, laboratory accreditation and intercomparison exercises

Analytical results are checked by experienced staff and discussed with senior scientists if questions arise.

DTU is accredited to testing for radioactivity by DANAK according to the international standard ISO 17025. The accreditation covers testing for certain non-gamma emitting radionuclides but not for radionuclides occurring in the environment and food in general.

DTU participate regularly in international intercomparisons on laboratory analyses of radionuclides.

CONCLUSIONS

This report shows the results of the environmental surveillance monitoring programme carried out at and around the Risø site in January-June 2017. The mean concentrations in air were: 0.29±0.28 μBq m^{-3} of ^{137}Cs, 2.92±0.92 mBq m^{-3} of ^{7}Be and 0.27±0.23 mBq m^{-3} of ^{210}Pb (±1 S.D.). The depositions by precipitation at Risø in the first half of 2017 were: 0.037±0.006 Bq m^{-2} of ^{137}Cs, 608±61 Bq m^{-2} of ^{7}Be, 12.7±1.1 Bq m^{-2} of ^{210}Pb and <0.9 kBq m^{-2} of ^{3}H. The average background dose rate (TLD) at Risø (Zone I) was measured as 51 nSv h^{-1} compared with 48 ± 3 nSv h^{-1} (±1 S.D.) in the four zones around Risø. None of the recorded levels of radioactivity and radiation have given rise to concern.

Center for Nuclear Technologies is Denmark's national competency center for nuclear technology. With roots in research in the peaceful use of nuclear power, DTU Nutech works with the applications of ionizing radiation and radioactive substances for the benefit of society.

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